Impact of medical waste incineration in the atmospheric
PCDD/F levels of Porto, Portugal

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Abstract

As a consequence of a monitoring program of a new municipal waste incinerator initiated in 1998, a large data-base of dioxin and furan concentrations in the atmosphere of the metropolitan area of Porto, in northern Portugal, has been collected. The existence of this data coincides with the shutdown in January 2001 of two medical waste incinerators that were under operation in the inner city of Porto. Dioxin emissions from these facilities were measured indicating emissions 100 to 1000 times larger than recent European Union directive limits. Data show that the shutdown of these two units had a clear effect on the improvement of air quality in the region that was observed either on the overall level of dioxins and furans or as in subtle alterations of the homolog pattern of these compounds in the atmosphere.
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1. Introduction

In 1997 the European Union (EU) concluded a first study for the development of an inventory of atmospheric emissions of polychlorinated dibenzo-p-dioxins (PCDD) and polychlorinated dibenzo–furans (PCDF) in Europe (Quass et al., 1997). According to that study, which reflected the emission situation for the period 1993–1995, substantial data gaps still existed for a number of potential and relevant dioxin emission sources. This study recommended additional research work that should focus on the development of measurement programs for all types of waste incinerators and diverse facilities in the metal production sector. Moreover, several countries had gathered little or none of their own relevant information up to that date. The lack of information was particularly noticeable in Portugal where no information about dioxin emissions measurements was available, despite the publication of a preliminary emission inventory for the metropolitan area of Porto (Coutinho et al., 1998) based on the application of emission factors.
As a consequence of this situation a second stage of the project was launched with the aim of reducing these uncertainties and to “catalyse” the implementation of corresponding national actions (Quass et al., 2004). The main objectives of this project were to carry out case studies on emission sources and, consequently, update the emission inventories on the basis of these results. Under this large framework, a monitoring program consisting of 13 samples covering the sectors of medical waste incineration, steel production and secondary aluminium and copper recovery was performed in Portugal within two measurement campaigns organized in December 1999 (Coutinho et al., 2000) and April 2000 (Coutinho et al., 2001a).

Parallel to this work on emissions, the construction of a municipal solid waste (MSW) incinerator in the region of Porto in 1998 led to the development of an external environmental monitoring program (Coutinho et al., 2001b). This monitoring program includes the regular sampling of heavy metals, and dioxins and furans in several matrices such as ambient air, water, sediments, soil, vegetables and diary products. This monitoring program is in regular operation currently. Since the beginning of this program a total of 93 ambient air samples were collected in several sites in the vicinity of the MSW incinerator providing an extensive characterization of dioxins and furans atmospheric levels in the metropolitan area of Porto, as well as information about the temporal trend of the atmospheric concentration of these compounds.

In the first trimester of 2001 the emission scenario in Porto experienced a significant reduction in the level of dioxins due to the shutdown of two medical waste incinerators responsible for an important fraction of the dioxin input to the atmosphere. This paper analyzes the reduction of the atmospheric levels of dioxins in the region of Porto as a consequence of the cessation of the operations of these facilities.

2. Experimental section

2.1. Emissions

The cooled probe method (EN 1948–1 of December 1996) was followed for the sampling procedure. In the application of this method, a partial volume of the flue gas is extracted via a glass tube from the flue gas duct and conducted into the collection system. The glass probe is mounted centrally in a water cooled titanium probe. A rapid cool down of the gaseous sample is achieved using cooling water. The collection system consists of a cartridge with two polyurethane foam absorption units. Polyurethane is a special sorbent with proven efficiency for the separation of organic substances. These foam units are installed on both sides of a plane filter with high collection efficiency. Connected to this absorption unit is an equipment group consisting of a drying tower, a gas pump and a gas volume meter. A sampling standard is added to the condensate flask before sampling. The flow of the sampled gas stream is adjusted to isokinetic conditions at the sampling nozzle.

2.2. Ambient air monitoring

The sampling apparatus for PCDDs/PCDFs in ambient air was carried out according to the German guideline VDI 3498 Part 2, 2002. The sampling apparatus consisted of a filter system, a mast, a suction pump, a gas volumeter and a timer. Particulate matter in the air was collected on a glass fiber filter. Filter-passing matter was collected on a polyurethane (PU) foam absorption filter. To monitor the effectiveness of sampling, a second polyurethane foam sampling unit was connected downstream. The reference volume is the volume of air that is drawn through the sampling device and measured with a gas volumeter during the sampling period (72 h).

The PCDD/PCDF samples which were deposited on the glass fibre filter and absorbed in the PU foam, were extracted and cleaned up from interfering components in a multistage separation process. They were quantified by gas chromatography/mass spectrometry, according to the German guideline VDI 3498 Part 1, 2002.

3. Results and discussion

3.1. Medical waste incinerators emissions

The city of Porto is the second largest city of Portugal with a metropolitan population of about
1,500,000 inhabitants. This region is served by two large general hospitals: The Hospital of S. João (HSJ) and the Hospital of S. António (HSA). These two hospitals have had their own facilities for the incineration of medical waste since the 1970s. Medical waste refers to any waste generated from the healthcare industry, including hospitals and medical laboratories. One of the major characteristics of medical waste is its heterogeneity. It includes anatomical waste, pathological waste, infectious waste, hazardous waste and other waste. Consequently, atmospheric emissions caused by the incineration of medical waste depend not only on the incineration conditions, but also on the waste characteristics.

Medical wastes must be classified according to their source, typology and risks factors associated with their handling, storage and ultimate disposal. Portuguese legislation classifies the waste stream produced through medical activities in four groups: Group I—similar to municipal wastes (MunW); Group II—non-hazardous medical waste that do not require specific treatment and can be considered similar to MunW; Group III—medical waste with biological risks that must be pre-treated before elimination as MunW; and Group IV—specific medical wastes with compulsory incineration. A recent work indicates that when waste collection follows usual segregation practices of HSA, Group I+II wastes...
represent 49% of the total, Group III 41% and Group IV only 10% on a weighted basis (Alvim-Ferraz and Afonso, 2003).

Incineration has been the most widely used treatment technology for the disposal of medical wastes. The major advantages are a significant reduction in the volume of material, and destruction of pathogens and hazardous organics. The main disadvantage is that incineration may emit trace amounts of unwanted pollutants such as PCDD and PCDF, usually in cities due to the typical location of hospitals (Lee and Huffman, 1996). That was the case of HSA and HSJ (see Fig. 1) that are located near two universities in Porto.

In 1999, stack PCDD/PCDF emission measurements were performed at these two hospitals incinerators in Porto (Coutinho et al., 2000). At the time, both hospitals were equipped with controlled air incinerators with two stage combustion. Waste was fed into the primary combustion chamber, which was operated with less than the full amount of air required for complete combustion. Under these sub-stoichiometric conditions, the waste was dried, heated and pyrolized. The non-volatile combustible portion was burned in the primary chamber. Moisture, volatiles and combustion gases from the primary chamber were mixed with air prior to entering the secondary combustion chamber where these combustible gases were ignited by a burner located near the entrance. Flue gases exiting the secondary chamber were sent directly to the stack (12 m high at HSJ and 30 m high at HSA) without any additional treatment. In the case of HSJ a heat recovery system was installed.

Emission data collected in 1999 from the two incinerators is summarized in Table 1. In order to analyze the results obtained, it is important to note that the EU emission limit value for new waste incinerators is 0.1 I-TEQ ng m$^{-3}$. Emissions resulting from the operation of these units were clearly above the EU limits: the emissions at HSA were 10 to 60 times higher than the limit, and HSJ emissions were three orders of magnitude above the limit. Taking into consideration the amount of waste burned during the sampling, it was possible to calculate an emission factor for PCDD/F that ranged between 30 and 200 I-TEQ µg of PCDD/F per ton of waste incinerated.

The HSA incinerator operated for a few hours, two days per week, for a total of about 416 h per year. On the other hand, HSJ collected waste from other hospitals and was in operation about 4800 h per year. As a consequence of this difference of the operational regime between the two incinerators, HSJ stands out as the major emitter of dioxins and furans to the atmosphere of Porto with a total of about 200 to 400 mg I-TEQ per year.

These results confirm previous studies published about medical waste treatment in Portugal that concluded that the incineration of medical waste without atmospheric pollutants control did not obey the legal emission limits, even when correct practices of operation and maintenance were used (Alvim-Ferraz and Pontes, 2000).

2,3,7,8-substituted PCDD/F congener profiles for the hospital waste incinerator emission samples are represented in Fig. 2. Dioxin congeners are depicted in dark grey and furans in light grey. Less chlorinated congeners are represented to the left of each set of emission bar graphs.

Table 1

<table>
<thead>
<tr>
<th>Sample</th>
<th>HSJ141299</th>
<th>HSJ151299</th>
<th>HSA071299</th>
<th>HSA171299</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stack temperature (°C)</td>
<td>185</td>
<td>191</td>
<td>435</td>
<td>430</td>
</tr>
<tr>
<td>Water content (%)</td>
<td>10.4</td>
<td>7.6</td>
<td>5.2</td>
<td>4.7</td>
</tr>
<tr>
<td>Oxygen (dry %)</td>
<td>14.8</td>
<td>14.0</td>
<td>17.6</td>
<td>16.3</td>
</tr>
<tr>
<td>Velocity (m/s)</td>
<td>1.8</td>
<td>1.5</td>
<td>12.0</td>
<td>13.2</td>
</tr>
<tr>
<td>Flow (dry Nm$^3$/h)</td>
<td>835</td>
<td>701</td>
<td>7330</td>
<td>8200</td>
</tr>
<tr>
<td>Concentration at 11% O$_2$ (I-TEQ ng/m$^3$)</td>
<td>125</td>
<td>102</td>
<td>1.03</td>
<td>5.88</td>
</tr>
<tr>
<td>Incinerated waste during sampling (kg)</td>
<td>2634</td>
<td>2796</td>
<td>485</td>
<td>700</td>
</tr>
<tr>
<td>Emission factor (I-TEQ µg/ton)</td>
<td>179</td>
<td>100</td>
<td>27</td>
<td>192</td>
</tr>
<tr>
<td>Hours of operation per year</td>
<td>4800</td>
<td>4800</td>
<td>416</td>
<td>416</td>
</tr>
<tr>
<td>Yearly emission (I-TEQ mg/yr)</td>
<td>380</td>
<td>220</td>
<td>1.4</td>
<td>9.3</td>
</tr>
</tbody>
</table>
Analysis of the individual congener pattern for the hospital waste incinerators samples shows a very strong homogeneity among the four samples. Remember that measurements were performed in two hospitals, with and without heat recovery. Congener profiles represented in Fig. 2 show a relative balance between dioxins and furans and the excess of the more chlorinated species.

As a consequence of the implementation of the national Strategic Plan for Medical Wastes (IR, 1998) these two incineration facilities were shutdown in January 2001. Since that time medical wastes are incinerated in a treatment facility with appropriate air pollutants control devices.

3.2. Ambient air

An ambient air monitoring program for the MSW incinerator in the region of Porto (see Fig. 1) was designed with the main objective of evaluating the effects on the surrounding area. In 1998, a set of two monitoring sites were selected in the suburbs of Porto (sites 1 and 3), in a radius of a few kilometres from the MSW complex, followed by another two in 1999 (sites 2 and 6). In August 1999, sampling ceased at the closest site (#3) to the facility. One of the sites located in downtown Porto (#6) was discontinued in February 2000. Regular monitoring continued at sites 1 and 2, with samples taken every 3 months until the present. Sites 4 and 5 were used during a specific field campaign organized in February 2002. Through the summer of 2004, more than 90 ambient air samples of PCDD/PCDF were taken during this 5-year period.

In this paper, in the computation of the total I-TEQ concentration calculated using I-TEF (International Toxicity Equivalency Factor), for those congeners below the detection limit the actual concentration was assumed to be half the detection limit.

The median concentration occurring during entire sampling period was of 130 fg I-TEQ m\(^{-3}\), with frequent samples above 400 fg I-TEQ m\(^{-3}\). Another ambient air monitoring program, developed in the Lisbon region, Portugal and described elsewhere (Coutinho et al., 2004), revealed a range of concentrations from 7.5 to 152 fg I-TEQ m\(^{-3}\). Fig. 3 illustrates several statistical parameters for all these samples and the ones collected in the Porto region. This figure shows that the 25th percentile for the Porto region (69 fg I-TEQ m\(^{-3}\)) is higher than the 75th percentile for the Lisbon region (41 fg I-TEQ m\(^{-3}\)).

The difference between the group of samples from Porto and from Lisbon cannot be explained by any natural characteristic of these regions. Both monitoring programs include territory with typical suburban...
land-use belonging to the metropolitan areas of Porto and Lisbon.

According to Lohmann and Jones (1998), PCDD/F concentrations for the total sum of TEQ are typically as follows: remote \(<10 \text{ fg I-TEQ m}^{-3}\); rural \(\sim20–50 \text{ fg I-TEQ m}^{-3}\); and urban/industrial \(\sim100–400 \text{ fg I-TEQ m}^{-3}\). Concentrations measured in Lisbon are comparable to those found in rural and uncontaminated urban areas: 83% of PCDD/PCDF concentrations in this region range from 10 to 100 fg I-TEQ m\(^{-3}\). On the other hand, in Porto 77% of samples collected are in the 40 to 400 fg I-TEQ m\(^{-3}\) range, approximately four times higher than the levels for the Lisbon region. Levels measured in Porto are consistent with data published for Barcelona, on the NE coast of the Iberian Peninsula, where the reported maximum concentrations were in the range of 600 to 800 fg I-TEQ m\(^{-3}\) (Abad et al., 2004).

Several studies performed in different airsheds (Hippelein et al., 1996; Fiedler et al., 1997) have shown that atmospheric levels of PCDD/PCDF follow a typical seasonal variation characterized by higher concentrations during winter when compared with typical summer concentrations. This seasonal pattern
can be explained by the intensification of the operation of diverse combustion sources during winter as well as by the more frequent presence of thermal inversion layers at the surface level during winter. These inversions cause a significant increase of atmospheric concentrations when pollutants are emitted at low levels below the thermal inversion layer.

To determine the seasonal behaviour of PCDD/F, two periods were considered, April–September and October–March, referred to as “summer” and “winter” in all figures presented in this paper. Fig. 4 shows the average PCDD/F concentration, expressed in I-TEQ, considering summer and winter periods in the region of Porto.

Prior to the winter of 2001–02, winter levels in Porto were approximately 3 to 4 times higher than summer levels. After then it is possible to observe a significant decrease of mean concentrations of atmospheric PCDD/PCDF. The mean value (37 fg I-TEQ m\(^{-3}\)) and concentration range (13–42 fg I-TEQ m\(^{-3}\)) for Summer 2002 are the lowest recorded in this region, followed by Summer 2003 (mean: 50 fg I-TEQ m\(^{-3}\), range: 9.8–172 fg I-TEQ m\(^{-3}\)). The decrease of atmospheric concentrations of PCDD/F was more evident during winter time: PCDD/F levels showed a reduction by a factor of 2, from average levels typically above 300 fg I-TEQ m\(^{-3}\) to values between 150 and 200 fg I-TEQ m\(^{-3}\).

For a more statistically significant approach, principal component analysis (PCA) was performed considering all seasonal periods since the beginning of PCDD/PCDF ambient air sampling in Porto. All data
were normalized so that \([\text{PCDD}] + [\text{PCDF}] = 1\), and average homologue contributions from each season, expressed as I-TEQ \(\text{m}^{-3}\), were used as inputs to the cluster analysis. PCA shows that data is subdivided in two clusters separating samples obtained before and after the shutdown of the medical waste incinerators (Fig. 5).

Homologue average profiles for each cluster were analysed in order to derive further indications on the possible dioxin sources (Fig. 6). The hepta- and octa-CDD and tetra-CDF have accounted for at least half of the total concentrations for the last few years. Since the shutdown of the medical waste incinerators, there has been an increased contribution of about 50% from the tetra-chlorinated species of furans, and a decrease to approximately one third of the hexa-chlorinated dioxins. Shutdown of the Hospital Waste incinerator at the beginning of 2001 resulted in a redistribution of homologue fractions, with an apparent increase in tetra-CDF species.

This relative increase helped to identify the contribution of one of the main sources of these congeners: the ferrous and non-ferrous metal industries operating in the region. A major seasonal pollution source of atmospheric PCDD/PCDF levels in the region may also be household wood burning for heating in winter. Burning woods at low temperatures, with low burning efficiency, plus burning of wood treated with chlorinated phenols (added as a preservative) woods may increase PCDD/PCDF emissions (Dyke et al., 1997).

4. Conclusions

Interpretation of temporal trends of atmospheric dioxin levels in the region of Porto shows the contribution that medical waste incinerators, without any air pollution control devices, might have in the deterioration of air quality in urban areas. The shutdown of two medical waste incinerators in Porto, Portugal caused a decrease in the atmospheric concentrations of dioxins and furans of about 50% in both winter and summer. Dioxins levels measured currently during the winter of approximately 200 fg I-TEQ \(\text{m}^{-3}\) reflect a relatively typical situation for a large urban area such as Porto. Nevertheless, these concentrations are significantly higher than levels monitored in a similar monitoring program in Lisbon and consequently deserve further research.

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